

# REQUEST FOR REPUBLICATION AND CORRECTED PUBLICATION PURSUANT TO 37 C.F.R. 1.221(b)

Docket No. C0003/7005C1

Applicant:

Karl Pichler and Carl Towns

Serial No:

10/615,924

Filed:

July 10, 2003

For:

ELECTROLUMINESCENT DEVICES WITH ELECTRODE PROTECTION

Examiner:

1BA 2879

Art Unit:

Publication No.: 2004/0023961 a1

Publication Date: February 3, 2005

Commissioner for Patents U.S. Patent and Trademark Office P.O. Box 1450 Alexandria, VA 22313-1450

Pursuant to 37 C.F.R. §1.221(b), this is a request for corrected publication of a material mistake made by the Office. This request is being filed within 2 months of the date of publication.

The publication error is the omission of claim 31 from the published application. The omitted claim is as follows:

31. A device as claimed in claim 30, wherein the thickness of the conductive oxide layer is between 10 and 500Å.

Applicant submits herewith a copy of the application papers as filed on July 10, 2003, with claims 1-31 (with postcards stamped by USPTO).

The error is believed to be material because the claims are the basis for provisional protection. Applicant requests a corrected publication.

Respectfully submitted,

Date: Opril 4, 2005

Therese A. Hendricks, Esq., Reg. No. 30,389

KUDIRKA & JOBSE, LLP Customer Number 021127

Tel: (617) 367-4600 Fax: (617) 367-4656

PATENT Customer No. 22,852 Attorney Docket No. 08513.7005-01

### IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

| In re Application of:  | )                                 |
|--|-----------------------------------|
| Pilcher et al.   | )<br>Group Art Unit: not assigned |
| Application No.: not assigned  | )<br>) Examiner: not assigned     |
| Filed: herewith  | )<br>)                            |
| For: ELECTROLUMINESCENT DEVICES WITH ELECTRODE PROTECTON               | )<br>)<br>)                       |
| Commissioner for Patents<br>P.O. Box 1450<br>Alexandria, VA 22313-1450 | ·                                 |
| Sir:   |                                   |

### PRELIMINARY AMENDMENT

Prior to the examination of the above application, please amend this application as follows:

### **IN THE SPECIFICATION:**

Amend the specification by inserting before the first line, the following new paragraph:

This is a continuation of Application No. 09/230,401, filed January 25, 1999, which is a national stage of PCT/GB97/02039 filed July 29, 1997, and which claims priority from GB 9615883.7 filed July 29, 1996, GB 9624707.7 filed November 28, 1996, GB 9703172.8 filed February 15, 1997 and GB 9619382.6 filed September 17, 1996, all of which are incorporated herein by reference, and from which priority is claimed.

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### IN THE CLAIMS:

Please cancel claims 1-25 and add the following new claims 26-31.

- 26. An organic electroluminescent device comprising:
  an anode formed of a positive charge carrier injecting material;
  a cathode formed of a negative charge carrier injecting material;
  a light emissive layer located between the anode and cathode; and
  a dielectric layer located between the light emissive layer and the anode.
- 27. A device as claimed in claim 26, wherein the thickness of the dielectric layer is between 10 and 500Å.
- 28. An organic electroluminescent device comprising:

  an anode formed of a positive charge carrier injecting material;

  a cathode formed of a negative charge carrier injecting material;

  a light emissive layer located between the anode and cathode; and

  a layer of carbon or amorphous silicon located between the light emissive

  layer and the anode.
- 29. A device as claimed in claim 28, wherein the thickness of the carbon or amorphous silicon layer is between 10 and 500Å.
- 30. An organic electroluminescent device comprising:

  an anode formed of a positive charge carrier injecting material;

  a cathode formed of a negative charge carrier injecting material;

  a light emissive layer located between the anode and cathode; and
  located between the light emissive layer and the anode, a layer of

  conductive oxide selected from the group consisting of tin oxide, zinc oxide, vanadium

  oxide, molybdenum oxide and nickel oxide.

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31. A device as claimed in claim 30, wherein the thickness of the conductive oxide layer is between 10 and 500Å.

### <u>REMARKS</u>

If there is any fee due in connection with the filing of this Preliminary

Amendment, please charge the fee to our Deposit Account No. 06-0916.

Respectfully submitted,

FINNEGAN, HENDERSON, FARABOW, GARRETT & DUNNER, L.L.P.

Dated:

July 9, 2003

Therese A. Hendricks

Reg. No. 30,389

FINNEGAN HENDERSON FARABOW GARRETT & DUNNER LLP

# Best Available Copy

# PLEASE STAMP TO ACKNOWLEDGE RECEIPT OF THE FOLLOWING:

In Re Application of: Pilcher et al.

Continuation of Application No.: 09/230,401

Group Art Unit: not yet assigned

Filed: herewith

Examiner: not yet assigned

For: ELECTROLUMINESCENT DEVICES WITH ELECTRODE PROTECTION

## PLEASE ACKNOWLEDGE RECEIPT OF THE FOLLOWING

Transmittal letter

Complete copy of prior application (09/230,401) as originally filed including drawings,
 Declaration and Assignment

3. Preliminary Amendment (3 pages)

4. Information Disclosure Statement (2 pages) with PTO Form 1149 (2 pages)

5. Check in the amount of \$750.00 for application filing fee

Dated 7/10/03

Docket No.: 08513.7005-01

TAH/jw - Mail Drop CAMB

JUL 10 2000 5

Dkt 07-11-03 pma

**BEST AVAILABLE COPY** 

# PLEASE ACCORD THIS NEW U.S. PATENT APPLICATION AN APPLICATION NUMBER AND FILING DATE

New U.S. Continuation Application for: ELECTROLUMINESCENT DEVICES WITH ELECTRODE PROTECTION

Inventors:

Pilcher et al.

APPLICATION NO.:

FILING DATE:

Dated: 7/10/03

Case Ref: 08513.7005-01 TAH/jw - Mail Drop No CAMB

### PLEASE STAMP TO ACKNOWLEDGE RECEIPT OF THE FOLLOWING:

In Re Application of: Pilcher et al.

Continuation of Application No.: 09/230,401

Group Art Unit: not yet assigned

Filed: herewith

Examiner: not yet assigned

For: ELECTROLUMINESCENT DEVICES WITH ELECTRODE PROTECTION

## PLEASE ACKNOWLEDGE RECEIPT OF THE FOLLOWING

- 1. Transmittal letter
- 2. Complete copy of prior application (09/230,401) as originally filed including drawings, Declaration and Assignment
- 3. Preliminary Amendment (3 pages)
- 4. Information Disclosure Statement (2 pages) with PTO Form 1149 (2 pages)
- 5. Check in the amount of \$750.00 for application filing fee

Dated 7/10/03

Docket No.: 08513.7005-01 TAH/jw - Mail Drop CAMB

PATENT Customer Number 22,852 Attorney Docket No. 08513.7005-01000

### IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Mail Stop Patent Application Commissioner for Patents P.O. Box 1450 Alexandria, VA 22313-1450

| Prior . | Applica     | tion Art Unit: 2812 Prior Application Examiner: S. Mulpuri   |
|---------|-------------|--|
| SIR:    | This is a   | a request for filing a   |
| § 1.53  | 3(b) of p   | tion Continuation-in-Part Divisional Application under 37 C.F.R. Dending prior Application No. 09/230,401 filed January 25, 1999 of Pichler CTROLUMINESCENT DEVICES WITH ELECTRODE PROTECTION.   |
| 1.      |             | Enclosed is a complete copy of the prior application (which is a national stage of PCT/GB97/02039 filed July 29, 1997, with an earliest priority date of July 29, 1996) including the oath or Declaration, Assignment and drawings as originally filed. I hereby verify that the attached papers are a true copy of prior Application No. 09/230,401 as originally filed in the U.S. Patent Office on January 25, 1999, which is incorporated herein by reference. |
| 2.      |             | Enclosed is a substitute specification under 37 C.F.R. § 1.125. The undersigned hereby verifies that no new matter is added in this substitute specification.  |
| 3.      |             | Enclosed is a Request for Non-Publication of Application and Certification Under 35 U.S.C. § 122(b)(2)(B)(i).  |
| 4.      | $\boxtimes$ | Cancel Claims 1-25.  |
| 5.      | $\boxtimes$ | A Preliminary Amendment adding new claims 26-31 is enclosed.   |
| 6.      |             | The filing fee is calculated on the basis of the claims existing in the prior application as amended at 4 and 5 above.   |
|         |             |  |

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| Basic Application Filing Fee                    |                  |   |       |                 |        | \$ | \$750.00 |
|---|------------------|---|-------|-----------------|--------|----|----------|
|   | Number of Claims |   | Basic | Extra<br>Claims |        |    |          |
| Total Claims                                    | 6                | - | 20    | 0               | x \$18 |    | 0        |
| Independent Claims                              | 3                | - | 3     | 3               | x \$84 |    | 0        |
| ☐ Presentation of Multiple Dep. Claim(s) +\$280 |                  |   |       |                 |        |    | 0        |
| Subtotal  |                  |   |       |                 |        | \$ | 750.00   |
| Reduction by 1/2 if small entity                |                  |   |       |                 |        |    | 0        |
| TOTAL APPLICATION                               | FILING FEE       |   |       | <del></del>     |        | \$ | 750.00   |

- 7. A check in the amount of \$750.00 to cover the filing fee is enclosed.
- 8. The Commissioner is hereby authorized to charge any fees which may be required including fees due under 37 C.F.R. § 1.16 and any other fees due under 37 C.F.R. § 1.17, or credit any overpayment during the pendency of this application to Deposit Account No. 06-0916.
- 9. Amend the specification by inserting before the first line, the following new paragraph:

This is a continuation of Application No. 09/230,401, filed January 25, 1999, which is a national stage of PCT/GB97/02039 filed July 29, 1997, which claims the benefit of GB 9615883.7 filed July 29, 1996, GB 9624707.7 filed November 28, 1996, GB 9703172.8 filed February 15, 1997, and GB 9619382.6 filed September 17, 1996 all of which are incorporated herein by reference.

- 10. New acceptable drawings are enclosed.
- 11. The prior application is assigned of record to: Cambridge Display Technology Limited
- 12. Priority of Applications GB 9615883.7 filed July 29, 1996, GB 9624707.7 filed November 28, 1996, GB 9703172.8 filed February 15, 1997 and GB 9619382.6 filed September 17, 1996, is claimed under 35 U.S.C. § 119. A certified copies

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 $\boxtimes$  are on file in the prior application. l is enclosed or 13. Small entity status is appropriate and applies to this application. 14.  $\boxtimes$ The power of attorney in the prior application is to at least one of the following: FINNEGAN, HENDERSON, FARABOW, GARRETT & DUNNER, L.L.P., Douglas B. Henderson, Reg. No. 20,291; Ford F. Farabow, Jr., Reg. No. 20,630; Arthur S. Garrett, Reg. No. 20,338; Donald R. Dunner, Reg. No. 19,073; Brian G. Brunsvold, Reg. No. 22,593; Tipton D. Jennings, IV, Reg. No. 20,645; Jerry D. Voight, Reg. No. 23,020; Laurence R. Hefter, Reg. No. 20,827; Kenneth E. Payne, Reg. No. 23,098; Herbert H. Mintz, Reg. No. 26,691; C. Larry O'Rourke, Reg. No. 26,014; Albert J. Santorelli, Reg. No. 22,610; Michael C. Elmer, Reg. No. 25,857; Richard H. Smith, Reg. No. 20,609; Stephen L. Peterson. Reg. No. 26,325; John M. Romary, Reg. No. 26,331; Bruce C. Zotter. Reg. No. 27,680; Dennis P. O'Reilley, Reg. No. 27,932; Allen M. Sokal. Reg. No. 26,695; Robert D. Bajefsky, Reg. No. 25,387; Richard L. Stroup. Reg. No. 28,478; David W. Hill, Reg. No. 28,220; Thomas L. Irving, Reg. No. 28,619; Charles E. Lipsey, Reg. No. 28,165; Thomas W. Winland, Reg. No. 27,605; Basil J. Lewris, Reg. No. 28,818; Martin I. Fuchs, Reg. No. 28,508; E. Robert Yoches, Reg. No. 30,120; Barry W. Graham, Reg. No. 29,924; Susan Haberman Griffen, Reg. No. 30,907; Richard B. Racine, Reg. No. 30,415; Thomas H. Jenkins, Reg. No. 30,857; Robert E. Converse, Jr., Reg. No. 27,432; Clair X. Mullen, Jr., Reg. No. 20,348; Christopher P. Foley, Reg. No. 31,354; John C. Paul, Reg. No. 30,413; David M. Kelly, Reg. No. 30,953; Kenneth J. Meyers, Reg. No. 25,146; Carol P. Einaudi, Reg. No. 32,220; Walter Y. Boyd, Jr., Reg. No. 31,738; Steven M. Anzalone, Reg. No. 32,095; Jean B. Fordis, Reg. No. 32,984; Roger D. Taylor, Reg. 28,992; Barbara C. McCurdy, Reg. No. 32,120; James K. Hammond, Reg. No. 31,964; Richard V. Burgujian, Reg. No. 31,744; J. Michael Jakes, Reg. No. 32,824; Thomas W. Banks, Reg. No. 32,719; Christopher P. Isaac, Reg. No. 32,616; Bryan C. Diner, Reg. No. 32,409; M. Paul Barker, Reg. No. 32,013; Andrew Chanho Sonu, Reg. No. 33,457; David S. Forman, Reg. No. 33,694; Vincent P. Kovalick, Reg. No. 32,867; James W. Edmondson, Reg. No. 33,871; Michael R. McGurk, Reg. No. 32,045; Joann M. Neth, Reg. No. 36,363; Gerson S. Panitch, Reg. No. 33,751; Cheri M. Taylor, Reg. No. 33,216; Charles E. Van Horn. Reg. No. 40,266; Linda A. Wadler, Reg. No. 33,218; Jeffrey A. Berkowitz. Reg. No. 36,743; Michael R. Kelly, Reg. No. 33, 921; James B. Monroe. Reg. No. 33,971; Doris Johnson Hines, Reg. No. 34,629; Lori Ann. Johnson, Reg. No. 34,498; R. Bruce Bower, Reg. No. 37,099; John Rissman, Reg. No. 33,764; Therese A. Hendricks, Reg. No. 30,389; Leslie I. Bookoff, Reg. No. 38,084; Michele C. Bosch, Reg. No. 40,524; Michael J. Flibbert, Reg. No. 33,234; Scott A. Herbst, Reg. No. 35,189;

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|   | ·   | Leslie A. McDonell, Reg. No. 34,872; Thalia V. Warnement, Reg. No. 39,064; Ronald A. Bleeker, Reg. No. 27,773; Kathleen A. Daley, Reg. No. 36,116; C. Gregory Gramenopoulos, Reg. No. 36,532; Anthony M. Gutowski, Reg. No. 38,742; Yitai Hu, Reg. No. 40,653; Lionel M. Lavenue; Reg. No. 46,859; Christine E. Lehman, Reg. No. 38,535; and Patrick J. Coyne, Reg. No. 31,821.  |
|---|---|--|
| 15.   |   | The original power appears in the original declaration of the prior application. A change of firm and address filed in the prior application is also enclosed.   |
| 16.   |   | Since the power does not appear in the original declaration, a copy of the power in the prior application is enclosed.   |
| 17.   |   | Please address all correspondence to Therese A. Hendricks, FINNEGAN, HENDERSON, FARABOW, GARRETT and DUNNER, L.L.P., 1300 I Street, N.W., Washington, D.C. 20005-3315, <b>Customer Number 22,852</b> .   |
| 18.   |   | Recognize as associate attorney  |
| 19.   |   | Also enclosed is   |
| applic<br>1999,<br>this ap<br>extens<br>neces | ation, in for the oplication is I sary fe | OR EXTENSION. If any extension of time is necessary for the filing of this including any extension in parent Application No. 09/230,401, filed May 10, purpose of maintaining copendency between the parent application and on, and such extension has not otherwise been requested, such an increby requested, and the Commissioner is authorized to charge es for such an extension to our Deposit Account No. 06-0916. A duplicate paper is enclosed for use in charging the deposit account. |
|   |   | FINNEGAN, HENDERSON, FARABOW,<br>GARRETT & DUNNER, L.L.P.  |
| Dated   | : 7/10                                    | 0/03 By: Thursday  |

Therese A. Hendricks Reg. No. 30,389

FINNEGAN HENDERSON FARABOW GARRETT & DUNNERLLP

PATENT Customer No. 22,852 Attorney Docket No. 08513.7005-01

### IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

| In re  | Application of:  | )                                 |
|--------|--|-----------------------------------|
| Pilche | er et al.  | )<br>Group Art Unit: not assigned |
| Appli  | cation No.: not assigned                                   | )<br>Examiner: not assigned       |
| Filed: | herewith   | )<br>)                            |
| For:   | ELECTROLUMINESCENT<br>DEVICES WITH ELECTRODE<br>PROTECTON  | )<br>)<br>)                       |
| P.O.   | nissioner for Patents<br>Box 1450<br>Indria, VA 22313-1450 |                                   |
| Sir:   |  |                                   |

### INFORMATION DISCLOSURE STATEMENT UNDER 37 C.F.R. § 1.97(b)

Pursuant to 37 C.F.R. §§ 1.56 and 1.97(b), applicant brings to the attention of the Examiner the documents listed on the attached PTO 1449. This Information Disclosure Statement is being filed within three months of the filing date of the above-referenced application.

Copies of the listed documents are of record in prior application no. 09/230,401, filed January 25, 1999, upon which applicant relies for the benefits provided in 35 U.S.C. § 120. Applicant respectfully requests that the Examiner consider the listed documents and indicate that they were considered by making appropriate notations on the attached form.

This submission does not represent that a search has been made or that no better art exists and does not constitute an admission that each or all of the listed

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documents are material or constitute "prior art." If the Examiner applies any of the documents as prior art against any claim in the application and applicant determines that the cited documents do not constitute "prior art" under United States law, applicant reserves the right to present to the office the relevant facts and law regarding the appropriate status of such documents.

Applicant further reserves the right to take appropriate action to establish the patentability of the disclosed invention over the listed documents, should one or more of the documents be applied against the claims of the present application.

If there is any fee due in connection with the filing of this Statement, please charge the fee to our Deposit Account No. 06-0916.

Respectfully submitted,

FINNEGAN, HENDERSON, FARABOW, GARRETT & DUNNER, L.L.P.

Dated:

July 9, 2003

y: The same

Therese A. Hendrick Reg. No. 30,389

FINNEGAN HENDERSON FARABOW GARRETT & DUNNER LLP

### INFORMATION DISCLOSURE CITATION

| Atty. Docket No. | 08513.7005-01  | Appln. No. | not yet assigned |  |
|------------------|----------------|------------|------------------|--|
| Applicant        | Pilcher et al. |            |                  |  |
| Filing Date      | herewith       | Group:     | not yet assigned |  |

|                      | U.S. PATENT DOCUMENTS |                |          |       |              |                            |
|----------------------|-----------------------|----------------|----------|-------|--------------|----------------------------|
| Examiner<br>Initial* | Document<br>Number    | Issue Date     | Name     | Class | Sub<br>Class | Filing Date If Appropriate |
|                      | *5,558,904 A          | Sept. 24, 1996 | Hsieh    | 427   | 66           | July 8, 1994               |
|                      | *5,247,190 A          | Sept. 21, 1993 | Friend   | 257   | 40           |                            |
|                      | *5,317,169            | May 31, 1994   | Nakano   | 257   | 40           | April 1, 1992              |
|                      | *5,766,515            | June 16, 1998  | Jonas    | 252   | 500          |                            |
|                      | *5,747,182            | May 5, 1998    | Friend   | 428   | 690          |                            |
|                      | *5,980,281            | Nov. 9, 1999   | Doi      | 428   | 690          |                            |
|                      | *5,821,002            | Oct. 13, 1998  | Danishi  | 428   | 690          |                            |
|                      | *6,002,206            | Dec. 14, 1999  | Harricon | 313   | 500          |                            |
|                      | *5,969,475            | June 19, 1999  | Friend   | 313   | 501          |                            |
|                      | *5,759,709            | June 2, 1998   | Doi      | 428   | 490          |                            |
|                      | *5,965,901            | Oct. 12, 1999  | Heeks    | 257   | 40           |                            |
|                      | *5,965,281            | Oct. 12, 2001  | Cho      | 428   | 690          |                            |
|                      | *6,416,885            | July 9, 2002   | Towns    | 428   | 690          |                            |

|                    | FOREIGN PATI        | ENT DOCUMENTS               |       |              |                          |
|--------------------|---------------------|-----------------------------|-------|--------------|--------------------------|
| Document<br>Number | Publication<br>Date | Country                     | Class | Sub<br>Class | Translation<br>Yes or No |
| <br>*EP 0443861 A2 | 28.08.91            | Europe                      |       |              |                          |
| *JP 6231881A       | 08.19.94            | Japan (English<br>Abstract) |       |              | •                        |
| *WO 95/24056       | 08.09.95            | PCT                         |       |              |                          |

| OTHER DOCUMENTS (Including Author, Title, Date, Pertinent Pages, Etc.)   |
|--|
| *Database WPI, Section Ch, Week 9343, Derwent Publications Ltd., London, GB; Class A26, AN 93-339976, XP002040832 & JP 05 247 460 A (Sumitomo Chem Co. Ltd.), 24 September 1993. |
| *Database WPI, Section EI, Week 9717, Derwent Publications Ltd., London, GB; Class U14, AN 97-185433, XP002040833 & JP 09 045 479 A (Hewlett Packard Co.), 14 February 1997.     |
| *Gruner et al. "Eission enhancement in single-layer conjugated polymer microcavitities", Journal of Appl. Phys. 80(1) July 1996  |

### INFORMATION DISCLOSURE CITATION

| Atty. Docket No. | 08513.7005-01   | Appln. No. not yet assigned   |
|------------------|---|---|
| Applicant        | Pilcher et al.  |   |
| Filing Date      | herewith  | Group: not yet assigned   |
|                  | OTHER DOCUMENTS (                                     | Including Author, Title, Date, Pertinent Pages, Etc.)   |
|                  | *Cho et al, Polymer light<br>the transparent anode" { | emitting diodes with polyethylene dioxythiopene polystyrene sulfonate as Synthetic metals, 87(1997) 171-174.                      |
| Examiner         | <del></del>   | Date Considered   |
| 1                |   | whether or not citation is in conformance with MPEP 609; draw line rmance and not considered. Include copy of this form with next |

Patent and Trademark Office - U.S. Department of Commerce

Form PTO 1449

2FORM PTO-1390 OFFICE (REV. 1094) U.S. PARTMENT OF COMMERCE PATENT AND TRADEMARK

TRANSMITTAL LETTER TO THE UNITED STATES
DESIGNATED/ELECTED OFFICE (DO/EO/US)

# ES

ATTORNEY'S DOCKET NUMBER C1043/7005

U.S. APPLICATION NO. (If known, see 37 CFR 1.5)

INTERNATIONAL APPLICATION NO. PCT/GB97/02039

INTERNATIONAL FILING DATE 29 July 1997 (29.07.97)

PRIORITY DATE CLAIMED 29 July 1996 (29.07.96)

| TITLE OF INVENTION                |                           |
|-----------------------------------|---------------------------|
| <b>ELECTROLUMINESCENT DEVICES</b> | WITH ELECTRODE PROTECTION |

APPLICANT(S) FOR DO/EO/US PICHLER, Karl and TOWNS, Carl

| Applicant herewith submits to the United States Designated/Elected Office (D | OO/EO/US) the | e following iten | ns and other information |
|--|---------------|------------------|--------------------------|
|--|---------------|------------------|--------------------------|

1. X This is a FIRST submission of items concerning a filing under 35 U.S.C. 371.

CONCERNING A FILING UNDER 35 U.S.C. 371

- This is a SECOND or SUBSEQUENT submission of items concerning a filing under 35 U.S.C. 371.
- 3. X This express request to begin national procedures (35 U.S.C. 371(f) at any time rather than delay examination until the expiration of the applicable time limit set in 35 U.S.C. 371(b) and PCT Articles 22 and 39(1).
- 1. X A proper Demand for International Preliminary Examination was made by the 19th month from the earliest claimed priority date.
- 5. X A copy of the International Application as filed (35 U.S.C. 371(c)(2)).
  - is transmitted herewith (required only if not transmitted by the International Bureau).
  - b. X has been transmitted by the International Bureau.
  - c. 

    is not required, as the application was filed in the United States Receiving Office (RO/US).
- 7. X Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371(c)(3)).
  - a.  $\square$  are transmitted herewith (required only if not transmitted by the International Bureau).
  - b. D have been transmitted by the International Bureau.
  - c. | have not been made; however, the time limit for making such amendments has NOT expired.
  - d. X have not been made and will not be made.
- 9. An oath or declaration of the inventor(s) (35 U.S.C. 371(c)(4)).
- 10. 

  A translation of the annexes to the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371(C)(5)).

### Items 11. To 16. Below concern document(s) or information included:

- 11. X An Information Disclosure Statement under 37 CFR 1.97 and 1.98.
- 12. 

  An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included.
- 13. X A FIRST preliminary amendment (with substitute specification, claims and abstract).
  - ☐ A SECOND or SUBSEQUENT preliminary amendment.
- 14. 

  A substitute specification (submitted as a first Preliminary Amendment).
- ☐ A change of power of attorney and/or address letter.
- 16. X Other items or information:

Mailed via Express Mailing Label No. EL056789709US

Published Application with Search Report

Post Card

Check No. 13199 for \$683.00

Express Mail Label No. EL056789709US

| U.S. APPLICATION NO. (If known  | LICATION NO. (If known, see 37 C.F.R. 1.5) INTERNATIONAL APPLICATION PCT/GB97/02039 |  |  | ATTORNEY'S DOCKET NUMBER<br>C1043/7005 |                 |
|---|---|--|--|--|-----------------|
| 17. X The following   | CALCULATIONS PTO USE ONLY   |  |  |  |                 |
| BASIC NATIONAL FI Search Report has be International prelim No international pre- but international sea Neither international fee (37 CFR 1.445) International prelim and all claims satisf   |   |  |  |  |                 |
|   |   | BASIC FEE AMOUNT   | =  | \$ 970.00                              |                 |
| Surcharge of \$130.00 for months from the earliest  | r furnishing the oath or d<br>claimed priority date (37                             | eclaration later than 20 7 CFR 1.492(e)).  | X 30   | \$                                     |                 |
| CLAIMS  | NUMBER FILED  | NUMBER EXTRA   | RATE   |  |                 |
| Total Claims  | 29 - 20 =   | 9  | X \$18.00                                    | \$162.00                               |                 |
| Independent Claims  | 6 - 3 =   | 3  | X \$78.00                                    | \$234.00                               |                 |
| MULTIPLE DEPENDE  | NT CLAIM(S) (if applic  | able)  | +\$260.00                                    | \$                                     |                 |
|   | TOTAL OF  | ABOVE CALCULATION  | ONS =  | \$1366.00                              |                 |
| Reduction by ½ for filin must also be filed (Note   | \$683.00  |  |  |  |                 |
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| Therese A. Hendrick<br>WOLF, GREENFIEL<br>600 Atlantic Avenue<br>Boston, Massachuse   | s<br>D & SACKS, P.C.  |  | Therese A. J. NAME  30,389  REGISTRATION NO. | Hendricks                              |                 |

### ATTORNEY'S DOCKET NO: C1043/7005

### IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Applicant:

Karl Pichler et al.

Serial No:

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Filed:

Herewith

For:

**ELECTROLUMINESCENT DEVICES WITH ELECTRODE** 

**PROTECTION** 

Assistant Commissioner for Patents Washington, D.C. 20231

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### PRELIMINARY AMENDMENT

Please amend the above application as follows.

### In The Claims

Please amend claims 6, 9, 11, 12, 13, 14, 15, 16, 19, 20, and 21 as follows:

- 6. (amended) A method as claimed in [any preceding claim] claim 1, wherein the anode protection layer is formed from polyethylene dioxythiophene/polystyrene sulphonate.
- 9. (amended) A method as claimed in [any preceding claim] claim 1, wherein a further layer is deposited between the anode protection layer and the light emissive layer to improve coating uniformity of the light emissive layer.
- 11. (amended) A method as claimed in [any preceding claim] claim 1, wherein a second light emissive layer is provided between the anode protection layer and the cathode.
- 12. (amended) A method as claimed [in any one of claims 1 to 5] claim 1, wherein when the anode protection layer is formed of carbon, tin oxide or silicon it is formed by sputtering or evaporating.

- 13. (amended) A method as claimed in [any preceding claim] claim 1, wherein the thickness of the anode protection layer is between 10 and 500Å.
- 14. (amended) A method according to [any preceding claim] claim 1, wherein the thickness of each of the anode and light emissive layer is between 500 and 2000Å.
- 15. (amended) A method according to [any preceding claim] claim 1, wherein the anode is formed of ITO.
- 16. (amended) A method according to [any preceding claim] claim 1, wherein the cathode is formed of aluminium or an aluminium alloy.
- 19. (amended) A device according to claim 17 [to] or 18, wherein the thickness of each of the anode and light emissive layer is between 500 and 2000Å.
- 20. (amended) A device according to [any of claims 17 to 19] claim 17, wherein the anode is formed of ITO.
- 21. (amended) A device according to [any of claims 17 to 20] claim 17, wherein the cathode is formed of aluminium or an aluminium alloy.

Respectfully Submitted,

Therese A. Hendricks

Reg. No. 30,389

Wolf, Greenfield & Sacks, P.C.

Theresel Dardruls

600 Atlantic Avenue

Boston, MA 02210-2211

(617)720-3500

Date: January 25, 1999 Docket No. C1043/7005

### WORLD INTELLECTUAL PROPERTY ORGANIZATION International Bureau



### INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

(51) International Patent Classification 6:

(11) International Publication Number:

WO 98/05187

H05B 33/10, 33/12, 33/14

(43) International Publication Date:

5 February 1998 (05.02.98)

(21) International Application Number:

PCT/GB97/02039

A1

(22) International Filing Date:

29 July 1997 (29.07.97)

(81) Designated States: CN, GB, JP, US, European patent (AT, BE, CH, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL,

PT, SE).

(30) Priority Data:

29 July 1996 (29.07.96) 9615883.7 GB 9619382.6 17 September 1996 (17.09.96) GB 9624707.7 GB

28 November 1996 (28.11.96)

9703172.8 15 February 1997 (15.02.97) **GB**  **Published** 

With international search report.

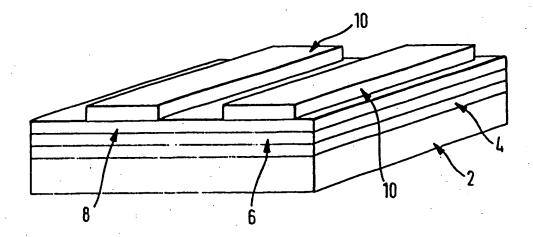
(71) Applicant (for all designated States except US): CAMBRIDGE DISPLAY TECHNOLOGY LIMITED [GB/GB]; 13 Station Road, Cambridge CB1 2JB (GB).

(72) Inventors; and

(75) Inventors/Applicants (for US only): PICHLER, Karl [AT/GB]; 3 Victoria Park, Cambridge CB4 3EJ (GB). TOWNS, Carl [GB/GB]; 193 Silver Street, Crown Cottage, Stanstead, Essex CM24 8HB (GB).

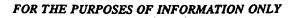
(74) Agents: DRIVER, Virginia, Rozanne et al.; Page White & Farrer, 54 Doughty Street, London WC1N 2LS (GB).

(54) Title: ELECTROLUMINESCENT DEVICES WITH ELECTRODE PROTECTION



### (57) Abstract

A method of manufacturing an electroluminescent device which has an anode (4) and a cathode (10) and arranged between the anode (4) and the cathode (10) a light emissive layer (8), also includes an anode protection layer (6) which protects the anode (10) against the effects of converting a precursor polymer to a semiconductive conjugated polymer which constitutes the light emissive layer (8). This has been found to increase the brightness and half-life of devices.



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### ELECTROLUMINESCENT DEVICES WITH ELECTRODE PROTECTION

### Field of the Invention

This invention relates to the construction of organic electroluminescent (EL) devices.

### Background of the Invention

Organic electroluminescent devices are made from materials that emit light when a suitable voltage is applied across electrodes deposited on either side of the material. One class of such materials is semiconductive conjugated polymers which have been described in our earlier Patent US 5,247,190, the contents of which are herein incorporated by reference. Poly(p-phenylene vinylene) [PPV], for instance, will emit light when positive and negative charge carriers are passed through the material by applying a voltage between two suitable electrodes. electroluminescent efficiency of these devices depends on the balancing of the electrons and holes that are injected into the device and meet to form electron/hole pairs, as well as on the efficiency with which these electron/hole pairs combine to radiate light, i.e. the photoluminescence efficiency (for example, see N.C. Greenham and R.H. Friend, Solid State Physics, 49, 1, 1995). Therefore it is of importance for an efficient device to have sufficiently high photoluminescence efficiency.

There are several approaches used for the processing of conjugated polymers. One approach uses a precursor polymer which is soluble and can therefore be easily coated by standard solution-based processing techniques (for example, spin-coating and blade-coating). The precursor is then converted in situ by suitable heat treatment to give the conjugated and insoluble polymer. Another approach uses directly soluble conjugated polymers which do not require a subsequent conversion stage. Depending on the specific application, one or other of the approaches might be relevant. The precursor polymers approach

can be especially important where subsequent processing might lead to damage of the polymer film if it were directly soluble such processing may be, for instance, coating with further polymer layers (for example, transport layers or emitting layers of different colour), or patterning of the top electrode. Converted precursor films also have better thermal stability which is of importance both during fabrication but also for the storage and operation of devices at high temperatures.

Where the precursor polymer is converted to the final form by elimination or modification of a solubilising group it is generally important that these by-products of the conversion process are removed from the film. It may also be important that they do not interact with the substrate during this process, for example if this causes harmful impurities to move into the film from the substrate thus affecting the performance (including luminescence efficiency and lifetime) of the electroluminescent device. We have observed, for instance, a quenching of the photoluminescence when precursor PPV polymers are converted on conductive oxide substrates such as indium tin oxide. This, we believe, may be caused by indium compounds being released into the PPV due to the reaction of one of the conversion by-products (for example, hydrogen halide) with the indium tin oxide.

In addition to the observation of quenching via the presence of impurities from the interaction of by-products with indium tin oxide during conversion, we have also observed detrimental effects due to the enhanced conversion of certain PPV copolymers. Such copolymers normally have limited conjugation lengths as compared to the homopolymer case. This normally leads to exciton confinement and therefore high photoluminescence and electroluminescence efficiencies. In this case, we believe that the indium compounds present in certain PPV copolymers films when converted on indium tin oxide can catalyse the elimination of groups designed to survive the conversion process.

### Summary of the Invention

The invention provides a device structure and a method of manufacture for an electroluminescent device that overcomes this problem.

According to one aspect of the invention there is provided a method of manufacturing an electroluminescent device comprising the steps of:

forming an anode of a positive charge carrier injecting material;

forming an anode protection layer on the anode of a protection material selected from the group comprising: polypyrroles and their derivatives; polythiophenes and their derivatives; polyvinylcarbazole (PVK); polystyrene; poly(vinyl pyridine); dielectric materials; carbon; amorphous silicon; non-indium containing conductive oxides including tin oxide, zinc oxide, vanadium oxide, molybdenum oxide and nickel oxide; and sublimed organic semiconductors;

forming a light emissive layer by converting a precursor to a polymer being a semiconductive conjugated polymer; and

forming a cathode of a negative charge carrier injecting material.

The anode protection layer has been found to be particularly valuable when the light emissive layer is a polymer which releases acidic by products (e.g. hydrogen halides) during the conversion from the precursor to the conjugated polymer.

Another aspect of the invention provides an electroluminescent device comprising:

an anode formed of a positive charge carrier injecting material;

an anode protection layer on the anode formed of a protection material selected from the group comprising: polypyrroles and their derivatives; polythiophenes and their derivatives; polystyrene; poly(vinyl derivatives; polyvinylcarbazole (PVK); polystyrene; poly(vinyl

pyridine); dielectric materials; carbon; amorphous silicon; nonindium containing conductive oxides including tin oxide, zinc oxide, vanadium oxide, molybdenum oxide, and nickel oxide; and sublimed organic semiconductors;

a light emissive layer formed of a semiconductive conjugated polymer; and

a cathode formed of a negative charge carrier injecting material.

The invention is particularly useful when the anode is formed of indium tin oxide (ITO). However other materials are suitable, such as tin oxide.

In one embodiment a layer of transparent conducting material deposited on glass or plastic forms the anode of the device. Examples of suitable anodes include tin oxide and indium tin Typical layer thicknesses are 500-2000A and sheet oxide. resistances are 10-100 Ohm/square, and preferably <30 Ohm/square. The converted precursor polymer can be, for instance, poly(pphenylene vinylene) [PPV] or a homopolymer or copolymer derivative of PPV. The thickness of this layer can be in the range 100-3000Å, preferably 500-2000Å and more preferably 1000-2000Å. The thickness of the precursor layer prior to conversion can be in the range 100-6000A for spin-coated layers and up to  $200\mu\text{m}$  for blade coating. The anode protection layer is chosen to act as a barrier against the conversion by-products of the precursor polymer, but also should not act as a barrier to the injection of holes from the anode into the emitting layer, where they combine with electrons injected from the cathode to radiate light. Conducting polymers are a general class of materials that can combine ease of processing, protection of the underlying electrode, and suitable hole transporting injecting and properties and are therefore good candidates. Thin layers of between 10-2000Å and preferably 10-500Å may be used and therefore the transparency of the layer can be high. Typical sheet resistances of these layers are 100-1000 Ohm/square, but can be as high as in excess of  $10^{10}\ \Omega/\text{squ}$ . Examples include conjugated

polymers that have been doped including polythiophenes, polyanilines, polypyrroles, and derivatives thereof. The cathode electrode is placed on the other side of the converted precursor material and completes the device structure. Furthermore, undoped conjugated polymers, as listed above, may also be used where the doping occurs in situ, by interaction with the conversion by-products during device manufacture.

The invention also provides use of an electrode protection layer in the manufacture of an organic light emitting device to protect an electrode of the organic light emitting device from the effects of conversion of a precursor into a light emitting semiconductive conjugated polymer, wherein the organic light emitting device comprises first and second electrodes with the light emitting polymer being located between them.

Thus, in another embodiment the electrode protection layer and the precursor polymer is deposited on the cathode, typically a material such as aluminium or an alloy of aluminium with a low work function element or any low work function element or alloy. In this case the protection layer will need to transport electrons, but may or may not need to be transparent. Again conducting polymers are suitable candidates as cathode protection layers. The anode electrode is placed on the other side of the converted precursor material and completes the device structure.

In yet another embodiment a protection layer to either the anode or cathode as described above is provided but where the protection layer is an undoped conjugated polymer but which has sufficient injection properties and transport mobilities for either holes or electrons depending on whether it is protecting the anode or cathode respectively. An example of such a protection layer would be a soluble PPV derivative or alternatively a precursor PPV or PPV derivative material. In the latter case, if the protection layer is much thinner than the electroluminescence layer, the by-products of the conversion process are more easily removed and therefore any interaction

with the electrode during conversion is reduced.

In yet another embodiment a protection layer to either the anode or cathode as described above is provided, but where the protection layer is an evaporated, sputtered, or reactively sputtered thin film which has sufficient injection properties and transport mobilities for either holes or electrons depending on whether it is protecting the anode or cathode respectively. An example of such a protection layer would be a thin layer of sputtered or evaporated carbon, a sputtered layer of amorphous silicon or non-indium containing conductive oxides including tin oxide, zinc oxide, vanadium oxide, molybdenum oxide, and nickel oxide, or a sublimed organic semiconductor layer.

In yet another embodiment a protection layer to either the cathode or anode as described above is provided, but where the protection layer is an undoped and non-conjugated polymer but which has sufficient injection properties and transport mobilities for either holes or electrons depending on whether it is protecting the anode or cathode respectively. An example would be polyvinyl carbazole which is a good hole transporting material but is not a conjugated polymer. Alternatively very thin layers of polymer materials which have relatively poor hole and electron mobilities may function as good electrode protectors without compromising the balance of electron and hole charge Examples would be polystyrene and poly(vinyl carriers. pyridine).

In yet another embodiment a protection layer to either the cathode or anode as described above is provided, but where the protection layer is a very thin inorganic dielectric which provides a barrier to the precursor conversion by-products, but which is thin enough that holes can tunnel through it when it is in contact and protecting the anode or electrons can tunnel through it when it is in contact and protecting the cathode.

The invention also provides a method of manufacturing an

electroluminescent device comprising the steps of:

forming an anode of a positive charge injecting material; forming a sacrificial anode protection layer over the anode; depositing a precursor to a semiconductive conjugated polymer on the sacrificial layer;

converting the precursor to a semiconductive conjugated polymer to form a light emitting layer, during which conversion step the anode protection layer protects the anode from the effects of the conversion and is itself consumed; and

forming a cathode of a negative charge injecting material.

Thus, in another embodiment a protection layer for either the anode or the cathode as described above is provided, but where the protection layer is a sacrificial layer. During the conversion process the sacrificial layer is etched away by the conversion by-products, the subsequence products of this interaction are chosen such that they do not interfere with the photoluminescence or electroluminescence efficiencies of the converted precursor conjugated polymers. Examples of such protection layers would include non-stoichiometric oxide films, such as silicon and aluminium oxides, the layer thickness being determined by the degree of interaction during the conversion process.

The invention also provides an organic light-emitting device, comprising:

an electrode;

an organic light-emissive layer formed from converted organic precursor; and

an electrode protection layer formed between the electrode and the light-emissive layer so as to protect the electrode during conversion of the organic precursor.

The invention also provides a method of manufacturing an organic light-emitting device, comprising the steps of:

depositing an electrode;

depositing an electrode protection layer over the electrode;

depositing a layer of an organic precursor for a lightemissive material; and

converting the organic precursor into the light-emissive material;

wherein the electrode protection layer protects the electrode during conversion of the organic precursor.

### Brief Description of the Drawings

Figures 1A to 1C are diagrams of an electroluminescent device incorporating an anode protection layer;

Figure 2 illustrates two conversion routes of a precursor to PPV;

Figure 3A and 3B are graphs illustrating the UV - vis spectra of PPV homopolymer respectively converted on quartz, indium tin oxide and an anode protection layer;

Figure 4 is a graph illustrating the UV - vis spectra of PPV copolymer converted on quartz, indium tin oxide and an anode protection layer; and

Figure 5 is a diagram illustrating the IR spectra of an acetate based copolymer converted on silicon, silicon with an indium layer, and silicon with an indium layer and protection layer.

For a better understanding of the present invention and to show how the same may be carried into effect reference will now be made by way of example to the above referenced drawings.

## Description of the Preferred Embodiments

Figure 1A illustrates a structure of an electroluminescent device. A substrate 2 formed of a transparent glass or plastics material is coated with a material constituting an anode 4 of the device. An anode protection layer 6 is located between the anode 4 and a light emitting layer 8. Cathode strips 10 are provided delineating with the anode 4 light emitting areas of the device. The operation of this device to emit light (without the anode

protection layer) is discussed in our preceding referenced Patent US 5,247,190 and will not be described further herein except to the extent that it is affected by the present invention.

### Embodiment I

A first embodiment is now described. Indium tin oxide constituting the anode 4 is deposited using either dc or rf sputtering techniques onto the polished glass substrate 2. Such substrates are available commercially. Soda lime glass with a thin silica barrier and an indium tin oxide layer of resistivity of 30 Ohm/square and transparency of about 85%, with a thickness of order 1500Å, can be used. A polythiophene based conducting polymer system is used as the anode protection layer 6. Polyethylene dioxythiophene/polystyrene sulphonate (PEDT/PSS @ 1:1.2 molar ratio) - which is available from Bayer AG, Leverkusen, Germany as Trial Product AI 4071. A 100Å film of the conducting polymer is spin-coated on the substrate. The EL layer 8 is formed by spin-coating a precursor polymer such as a homopolymer PPV. With this precursor polymer the solubilising group that is removed during conversion at 150°C in nitrogen for 4 hours is tetrahydrothiophene, and the counter-ion to the thiophene salt is bromide. Another by-product is therefore hydrogen bromide which readily attacks ITO and can cause the release of detrimental products into the film which quenches the photoluminescence. The conversion by-products of the PPV-based precursor are indicated in Figure 2 where a=0, a'=0.

Without the anode protection layer, initial measurements of PL efficiency of the PPV material were reduced from about 13% to, at best, about 0.7% following the thermal conversion process. Further measurements established that the PL efficiency may be in the range 10% down to about 2-3%. Initial measurements with the anode protection layer indicated a PL efficiency of ~3%. Subsequent work has shown that this can be increased to ~5%. After the conversion a suitable cathode material, calcium for instance, is deposited on top of the conjugated polymer 8 and

patterned to form strips 10. After that, contacting and encapsulation with epoxy/glass were immediately performed in a glove box. Devices made with the protector layer typically have significantly improved electroluminescence efficiency compared to the devices without the protector layer 6.

### Embodiment II

Another specific embodiment is now described. The initial steps are the same as embodiment I up to formation of the EL layer. In this embodiment, a precursor to an acetate-based PPV copolymer This material has a very high photoluminescence is deposited. (PL) efficiency, where the solubilising group that is removed during conversion is tetrahydrothiophene, and the counter-ion to the thiophene salt is bromide. Another by-product is therefore hydrogen bromide which readily attacks ITO and can cause the release of detrimental products into the film which quenches the photoluminescence and causes enhanced conversion. anode protection layer 6, the PL efficiency of the PPV material is dramatically reduced from about 50-60% to, at best about 7% following the thermal conversion process (150°C in nitrogen for 4 hours as before). However, with the protector layer a PL efficiency of ~22% is obtained following conversion. Figure 2 shows the conversion system, where  $a\neq 0$ ,  $a'\neq 0$ . conversion a suitable cathode material, calcium for instance, is deposited on top of the conjugated polymer.

Table 1 illustrates the photoluminescent efficiencies for embodiments I and II, in the final column of Table 1. The first and second columns of Table 1 illustrate values for the photoluminescence efficiency in situations where the precursor layer is spin-coated onto quartz and indium tin oxide respectively without the use of the anode protection layer. Table 1A shows equivalent figures resulting from what we believe are more accurate measurements with a better statistical base.

The copolymer referred to in this case was measured initially to

contain ~20 mol.% of the acetate function. Subsequent measurements which we believe to be more accurate indicate a content of ~40 mol.% of the acetate function. Modification of the copolymer acetate level has led to photoluminescence efficiencies of about 30% when converted on ITO with the PEDT/PSS protection layer.

Figures 3 to 5 show that protection of the PPV copolymer is also brought about minimising the enhanced conversion with the ITO protection layer. Figure 3A illustrates measurements taken from structures having differing layer thicknesses. Figure 3B shows the situation where a common layer thickness is used. Figure 3B illustrates that the UV - vis spectra show little change in the homopolymer case irrespective of the substrate used. Figure 4 shows that there is an enhanced red shift for the acetate based copolymer when converted on ITO. In addition, there is an absorption peak at 1737 cm-1 in the IR spectra which is assigned to the acetate carbonyl absorption. The relative intensity of this can be compared with other peaks in the spectrum, such as the absorption at 1517 cm-1 which originates in the aromatic constituents of the polymer. The ratio of the intensities of the two peaks therefore gives a measure of the relative quantities of the acetate function. Table 2 shows that this ratio (acetate:aromatic) is significantly reduced when the conversion is carried out on silicon with an indium layer. interpret these results as enhanced conversion of the acetate based copolymer by indium compounds from the silicon substrate with indium layer and this process is reduced by the presence of protection layers. Relative photoluminescence efficiencies are detailed in Tables 1A and B. The device performance of the systems including the protection layer may be summarised as 100cd/m2 starting brightness, efficiency of 0.2-0.6 lm/W and up to 2 lm/W, with a half-life of brightness (at constant current or constant voltage drive) of 10-100 hours, and up to 2000 hours.

### Embodiment III

specific embodiment is now described. Another this embodiment, the production steps are the same for Embodiment II except that the polyethylene dioxythiophene/polystyrene sulphonate material which is used as the anode protection layer has been optimised to give beneficial lifetime performance by increasing the PSS content. Thus, the material now has a 1:5 molar ratio PEDT/PSS. The device performance of these system may be summarised as 100 cd/m2 starting brightness; efficiency of 0.3-1.2 lm/W, and up to 2 lm/W with a half-life of ~500 hours and up to 2000 hours.

### Embodiment IV

In the case of Embodiment III, we have observed a detrimental interaction between the PEDT/PSS protection layer (@ 1:5 molar ratio) with the PPV precursor solution. We believe this is because of dissolution of the PEDT/PSS layer in the PPV precursor solution and this can lead to non-uniform emission in the final For example, if the PPV is spin-coated on top of the PEDT/PSS film during device fabrication then a circular nonuniformity is observed at the PEDT/PSS-PPV interface after conversion. We have overcome this problem by spin-coating a thin poly(vinyl pyridine) (PVP) film (Figure 1B - reference 7) on top of the PEDT/PSS layer before the PPV precursor solution is As is well understood, commercially available PVP includes a component of polystyrene, typically 10%, to render it soluble. Hence, a 100Å film of the PEDT/PSS system is deposited as described above and following this a thin PVP film is spincoated from a 0.1% w/v solution in methanol. The rest of the device is manufactured in the normal way and characteristics as outlined above are obtained (i.e. 100 cd/m2 initial brightness, 0.3-1.2 lm/W efficiency, with a half-life of ~500 hours). However, the emission uniformity is greatly improved. As the PVP acts as a barrier between the PEDT/PSS system and the PPV, this approach can also be used to pattern this ITO protection layer.

### Embodiment V

A further specific embodiment is now described and relates to the fabrication of such devices. A sheet of ITO coated glass is taken and cleaned. The dimensions of the ITO-coated glass may be from 12\*12 mm to much greater than 80\*80 mm. The PEDT/PSS ITO protection layer is then spin-coated onto the substrate to a thickness of ~100Å. Following this the PPV precursor solution is blade-coated onto the PEDT/PSS layer at a wet film thickness of 100  $\mu m$  at a precursor solution concentration of 0.4-0.5% solid content. In this case the device uniformity is superior to that obtained when the PPV precursor is spin-coated. Alternatively, a double layer PPV device may be blade-coated such that each layer is ~500-700Å thick and a short conversion (~20 minutes at 150°C) is carried out before deposition of the second layer (reference 9 in Figure 1C). After conversion the final conversion the PPV film obtained is ~1000-1400A thick. case beneficial effects are observed with respect to device efficiency and gross uniformity. A suitable cathode is then deposited and the device is connectorised.

### Embodiment VI

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In another embodiment, a glass substrate is coated with indium tin oxide in the manner described above. Then, PVP was dissolved in methanol to a concentration of 0.1%, prefiltered to 1 micron pore size and coated onto the indium tin oxide to a thickness of about 100Å. Then, the PPV precursor discussed above with reference to Embodiment I is spincoated on top and converted at 150°C in nitrogen for 4 hours to render a layer of PPV of about 1000Å thickness. The device was then stored in a desiccator for 48 hours before a cathode formed from an aluminium/lithium alloy was sputtered on top.

### Embodiment VII

This embodiment was formed in the same manner as Embodiment VI,

except that the anode protection layer was formed of polyvinylcarbazole (PVK) dissolved in THF to a concentration of 0.1%.

### Embodiment VIII

This embodiment was formed in the same manner as Embodiments VI and VII except that the anode protection layer was formed of polystyrene dissolved in THF to a concentration of 0.1%.

### Embodiment IX

This embodiment was formed in the same manner as Embodiments VI, VII and VIII except that the anode protection layer was formed of poly(vinyl pyridine) dissolved in methanol to a concentration of 0.1%.

### Embodiment X

In another embodiment, the device is manufactured according to Embodiment II, but the cathode is formed of a lithium/aluminium alloy instead of calcium. For instance a lithium/aluminium alloy containing up to 10% by weight Li, is sputtered on top of the conjugated polymer to a thickness of  $10\text{\AA}\text{-}1\mu\text{m}$  and preferably ~1200Å. The Li/Al alloy targets are commercially available and can typically contain ~2.5% by weight Li. Other stabilising elements such as Zr, Mg, Cu may also be present. Devices made with the protector layer and the lithium based cathode have significantly improved electroluminescence efficiencies compared to the devices without the protector layer and using say calcium electrode.

Thus, the various embodiments described above of the present invention each provide a multilayer electroluminescent device incorporating a converted precursor polymer as the emitting layer and an electrode protecting layer placed between the converted precursor polymer and the underlying electrode and which acts to

protect the electrode during the precursor conversion process. At least one other layer is present one of which is the second electrode.

The embodiments described above are illustrative of a method of manufacture of an electroluminescence device wherein a precursor to a conjugated polymer material is deposited on a substrate on which has previously been deposited both an electrode layer and subsequently an electrode protection layer. The precursor is then converted to the final conjugated polymer form before deposition of a subsequent layer or layers at least one of which is the second electrode.

TABLE 1
TYPICAL PHOTOLUMINESCENCE EFFICIENCY (%) MEASUREMENTS

| Polymer Type | PL eff/Quartz | PL eff/ITO | PL eff/Protection<br>Layer /ITO |
|--------------|---------------|------------|---------------------------------|
| Homopolymer  | 13.2          | 0.7        | 3                               |
| Copolymer    | 56            | 6.8        | 22                              |

TABLE 1A

IMPROVED PHOTOLUMINESCENCE EFFICIENCY (%) MEASUREMENTS

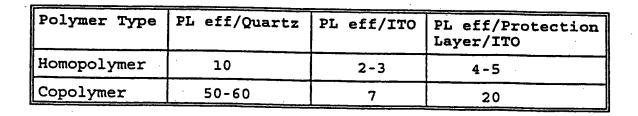




TABLE 2

1737/1517 cm-1 Ratios (Acetate:Carbonyl) from IR spectra

| Substrate                            | 1737/1517 cm-1<br>Ratio* |  |
|--------------------------------------|--------------------------|--|
| Inert(Si)                            | 1.1                      |  |
| Si with indium layer with protection | 1                        |  |
| Si with indium layer                 | 0.3                      |  |

#### CLAIMS:

1. A method of manufacturing an electroluminescent device comprising the steps of:

forming an anode of a positive charge carrier injecting material;

forming an anode protection layer on the anode of a protection material selected from the group comprising: polypyrroles and their derivatives; polythiophenes and their derivatives; polyvinylcarbazole (PVK); polystyrene; poly(vinyl pyridine); dielectric materials; carbon; amorphous silicon; non-indium containing conductive oxides including tin oxide, zinc oxide, vanadium oxide, molybdenum oxide and nickel oxide; and sublimed organic semiconductors;

forming a light emissive layer by converting a precursor to a polymer being a semiconductive conjugated polymer; and

forming a cathode of a negative charge carrier injecting material.

- 2. A method as claimed in claim 1, wherein the semiconductive conjugated polymer is selected from a class of polymers which release acidic by products during the conversion from the precursor polymer to the conjugated polymer.
- 3. A method as claimed in claim 1 or 2, wherein the anode comprises a transparent conducting layer deposited on a substrate of a glass or plastics material.
- 4. A method as claimed in claim 1 or 2, wherein the light emissive layer is formed from a homopolymer of poly(p-phenylene vinylene) (PPV).
- 5. A method as claimed in claim 1 or 2, wherein the light emissive layer is formed from an acetate based copolymer of PPV.
- 6. A method as claimed in any preceding claim, wherein the anode protection layer is formed from polyethylene

dioxythiophene/polystyrene sulphonate.

- 7. A method as claimed in claim 6, wherein the molar ratio of the anode protection layer is 1:1.2 PEDT/PSS.
- 8. A method as claimed in claim 6, wherein the molar ratio of the anode protection layer is 1:5 PEDT/PSS.
- 9. A method as claimed in any preceding claim, wherein a further layer is deposited between the anode protection layer and the light emissive layer to improve coating uniformity of the light emissive layer.
- 10. A method as claimed in claim 9, wherein the further layer comprises poly(vinyl pyridine) (PVP).
- 11. A method as claimed in any preceding claim, wherein a second light emissive layer is provided between the anode protection layer and the cathode.
- 12. A method as claimed in any of claims 1 to 5, wherein when the anode protection layer is formed of carbon, tin oxide or silicon it is formed by sputtering or evaporating.
- 13. A method as claimed in any preceding claim, wherein the thickness of the anode protection layer is between 10 and 500Å.
- 14. A method according to any preceding claim, wherein the thickness of each of the anode and light emissive layer is between 500 and 2000Å.
- 15. A method according to any preceding claim, wherein the anode is formed of ITO.
- 16. A method according to any preceding claim, wherein the cathode is formed of aluminium or an aluminium alloy.

17. An electroluminescent device comprising:

an anode formed of a positive charge carrier injecting material;

an anode protection layer on the anode formed of a protection material selected from the group comprising: polypyrroles and their derivatives; polythiophenes and their derivatives; polyvinylcarbazole (PVK); polystyrene; poly(vinyl pyridine; dielectric materials; carbon; amorphous silicon; non-indium containing conductive oxides including tin oxide, zinc oxide, vanadium oxide, molybdenum oxide and nickel oxide;

a light emissive layer formed of a semiconductive conjugated polymer; and

a cathode formed of a negative charge carrier injecting material.

- 18. A device as claimed in claim 17, wherein the thickness of the anode protection layer is between 10 and 500Å.
- 19. A device according to claim 17 to 18, wherein the thickness of each of the anode and light emissive layer is between 500 and 2000Å.
- 20. A device according to any of claims 17 to 19, wherein the anode is formed of ITO.
- 21. A device according to any of claims 17 to 20, wherein the cathode is formed of aluminium or an aluminium alloy.
- 22. A method of manufacturing an electroluminescent device comprising the steps of:

forming an anode of a positive charge injecting material; forming a sacrificial anode protection layer over the anode; depositing a precursor to a semiconductive conjugated polymer on the sacrificial layer;

converting the precursor to a semiconductive conjugated polymer to form a light emitting layer, during which conversion step the anode protection layer protects the anode from the

effects of the conversion and is itself consumed; and forming a cathode of a negative charge injecting material.

- 23. Use of an electrode protection layer in the manufacture of an organic light emitting device to protect an electrode of the organic light emitting device from the effects of conversion of a precursor into a light emitting semiconductive conjugated polymer, wherein the organic light emitting device comprises first and second electrodes with the light emitting polymer being located between them.
- 24. An organic light-emitting device, comprising: an electrode;

an organic light-emissive layer formed from converted organic precursor; and

an electrode protection layer formed between the electrode and the light-emissive layer so as to protect the electrode during conversion of the organic precursor.

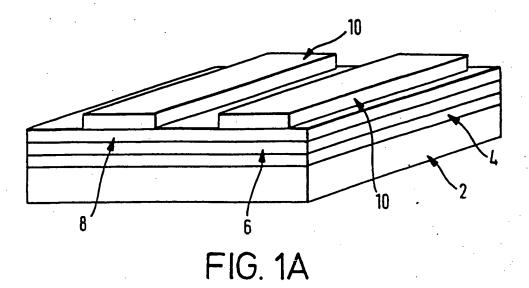
25. A method of manufacturing an organic light-emitting device, comprising the steps of:

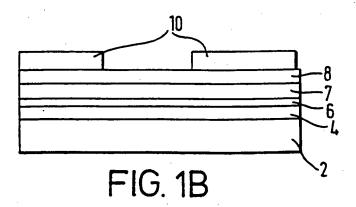
depositing an electrode;

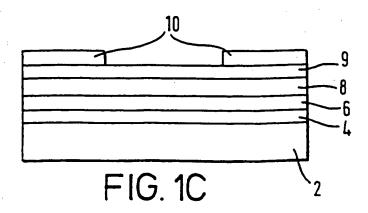
depositing an electrode protection layer over the electrode; depositing a layer of an organic precursor for a lightemissive material; and

converting the organic precursor into the light-emissive material;

wherein the electrode protection layer protects the electrode during conversion of the organic precursor.



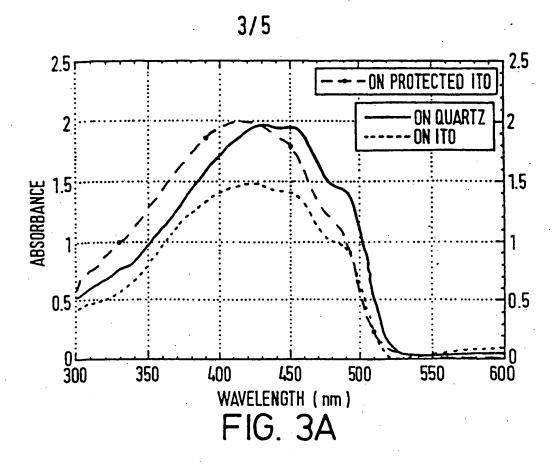


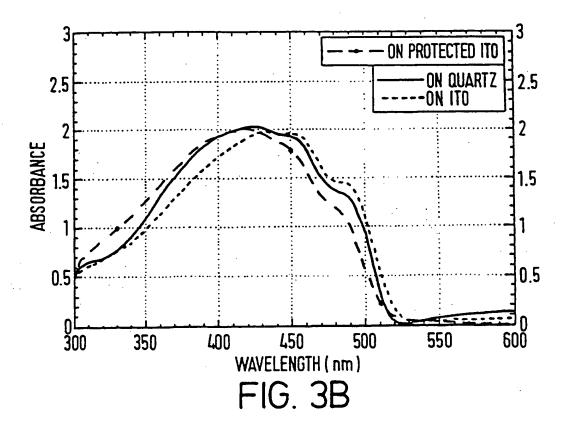


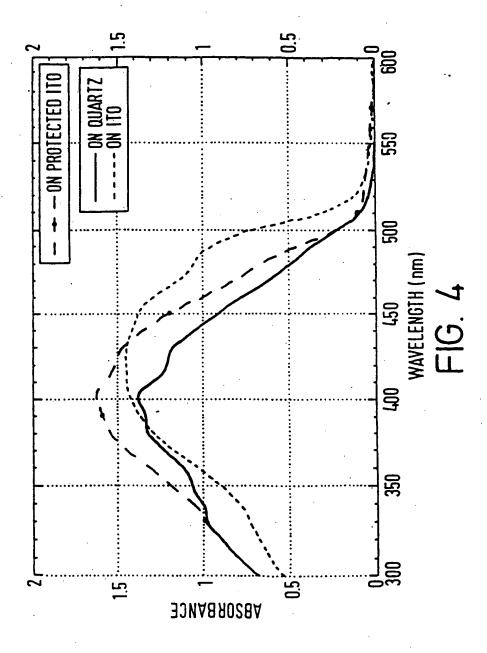
$$XH_{2}C \bigcirc CH_{2}X \qquad xs \bigcirc S \qquad \downarrow 2X^{-} \\ p-Xylenebis \{tetrahydrothiopheniumbromide\} \\ Monomer \\ \downarrow SH_{2}C \bigcirc CH_{2}^{+}S \bigcirc CH_{2$$

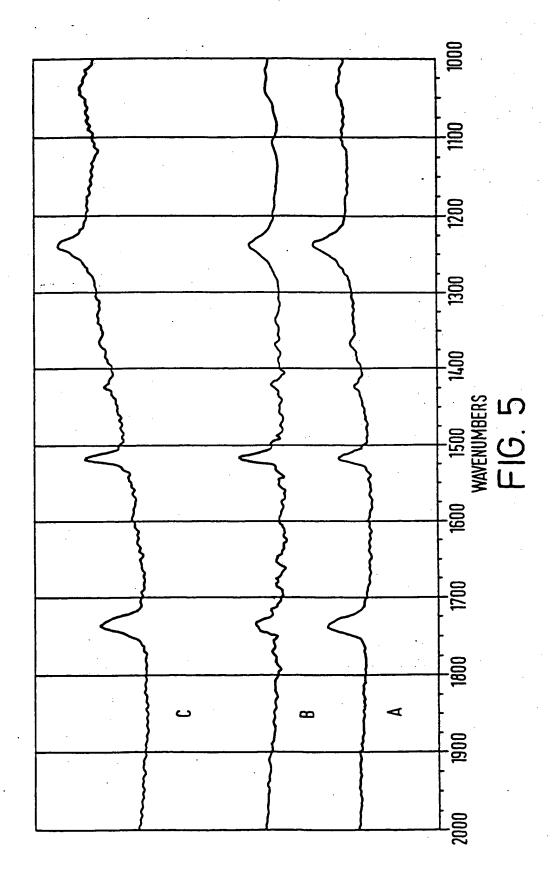
CONVERTED PPV COPOLYMER
For the homopolymer 
$$n^1 > a^1$$
,  $n > a = 0$ 
For the copolymer  $n^1 > a^1$ ,  $n > a = (a, a^1 \neq 0)$ 

F1G. 2













### INTEL AMONAL SEARCH REPORT

PCT/GB 97/02039

A. CLASSIFICATION OF SUBJECT MATTER
IPC 6 H05B33/10 H05B33/12 H05B33/14

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols) IPC 6 H05B

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

| C. DOCUMENTS | CONSIDERED TO BE RELEVANT |
|--------------|---------------------------|
|--------------|---------------------------|

European Patent Office, P.B. 5818 Patentiaan 2 Nr. - 2280 HV Billingiille

| Category * | Citation of document, with indication, where appropriate, of the relevant passages   | Relevant to claim No. |
|------------|--|-----------------------|
| A,P        | US 5 558 904 A (B.R.HSIEH & AL) 24<br>September 1996<br>see column 6, line 1-38; claims 1-14   | 1-5,<br>12-25         |
| A          | DATABASE WPI Section Ch, Week 9343 Derwent Publications Ltd., London, GB; Class A26, AN 93-339976 XP002040832 & JP 05 247 460 A (SUMITOMO CHEM CO LTD), 24 September 1993 see abstract | 1-5,<br>12-25         |
|            | -/   |                       |
|            |  | ł                     |

| Further documents are listed in the continuation of box C.  | X Patent family members are listed in annex.   |
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| Date of the actual completion of the international search  7 October 1997   | Date of mailing of the international search report  17.10.97   |
| Name and mailing address of the ISA   | Authorized officer   |

1

# INTEP AONAL SEARCH REPORT

| C.(Continua | tion) DOCUMENTS CONSIDERED TO BE RELEVANT   | PCT/GB 9 | 7/02039               |
|-------------|---|----------|-----------------------|
| Category °  | Citation of document, with indication, where appropriate, of the relevant passages  |          | Relevant to claim No. |
| A,P         | DATABASE WPI Section EI, Week 9717 Derwent Publications Ltd., London, GB; Class U14, AN 97-185344 XP002040833 & JP 09 045 479 A (HEWLETT-PACKARD CO), 14 February 1997 see abstract |          | 1-5                   |
|             | US 5 247 190 A (R.H.FRIEND) 21 September 1993 cited in the application see the whole document   |          | 1-5,<br>12-25         |
|             |   |          |                       |
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## MAL SEARCH REPORT

PCT/GB 97/02039

| Patent docu<br>cited in search |      | Publication date | Patent family member(s) | Publication date |
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|                                |      |                  | AU 5428590 A            | 16-11-90         |
|                                |      | ·                | CA 2030785 A            | 21-10-90         |
|                                | •    | •                | DE 69016345 D           | 09-03-95         |
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|                                |      |                  | ES 2070320 T            | 01-06-95         |
|                                |      |                  | WO 9013148 A            | 01-11-90         |
|                                |      |                  | HK 24597 A              | 27-02-97         |
|                                |      |                  | JP 4500582 T            | 30-01-92         |
|                                |      |                  | US 5399502 A ,          | 21-03-95         |

### INTEL AMONAL SEARCH REPORT

PCT/GB 97/02039

A CLASSIFICATION OF SUBJECT MATTER
IPC 6 H05B33/10 H05B33/12 H05B33/14

According to International Patent Classification (IPC) or to both national classification and IPC



#### B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols) IPC 6 H95B

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

| Category * | Citation of document, with indication, where appropriate, of the relevant passages   | Relevant to claim N |
|------------|--|---------------------|
| A,P        | US 5 558 904 A (B.R.HSIEH & AL) 24<br>September 1996<br>see column 6, line 1-38; claims 1-14   | 1-5,<br>12-25       |
| A          | DATABASE WPI Section Ch, Week 9343 Derwent Publications Ltd., London, GB; Class A26, AN 93-339976 XP002040832 & JP 05 247 460 A (SUMITOMO CHEM CO LTD), 24 September 1993 see abstract | 1-5,<br>12-25       |
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| X Further documents are listed in the continuation of box C.   | X Patent family members are listed in annex.   |
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| Date of the actual completion of the international search  | Date of mailing of the international search report   |
| 7 October 1997   | 17.10.97   |
| Name and mailing address of the ISA  | Authorized officer   |

European Patent Office, P.B. 5818 Patentiaan 2 NL - 2280 HV Rijswijk

## INTERNATIONAL SEARCH REPORT

PCT/GB 97/0203

| Category * | Citation of document with indication where are  |                       |
|------------|---|-----------------------|
|            | Citation of document, with indication, where appropriate, of the relevant passages  | Relevant to claim No. |
| A, P       | DATABASE WPI Section EI, Week 9717 Derwent Publications Ltd., London, GB; Class U14, AN 97-185344 XP002040833 & JP 09 045 479 A (HEWLETT-PACKARD CO), 14 February 1997 see abstract | 1-5                   |
| \          | US 5 247 190 A (R.H.FRIEND) 21 September  | 1-5,                  |
| :          | 1993 cited in the application see the whole document  | 12-25                 |
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ation on patent family members

|        | Application M |
|--------|---------------|
| PCT/GB | 97/02039      |

| Patent document cited in search report | Publication<br>date | Patent family<br>member(s) | Publication date |
|--|---------------------|----------------------------|------------------|
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| US 5247190 A                           | 21-09-93            | AT 117834 T                | 15-02-95         |
|  |                     | AU 626415 B                | 30-07-92         |
|  |                     | AU 5428590 A               | 16-11-90         |
|  |                     | CA 2030785 A               | 21-10-90         |
| ,                                      |                     | DE 69016345 D              | 09-03-95         |
| •                                      |                     | DE 69016345 T              | 24-05-95         |
|  |                     | EP 0423283 A               | 24-04-91         |
|  |                     | ES 2070320 T               | 01-06-95         |
|  |                     | WO 9013148 A               | 01-11-90         |
| 4                                      |                     | HK 24597 A                 | 27-02-97         |
|  |                     | JP 4500582 T               | 30-01-92         |
|  |                     | US 5399502 A .             | 21-03-95         |

#### DECLARATION FOR PATENT APPLICATION



As a below named inventor, I hereby declare that:

My residence, post office address and citizenship are as stated below next to my name.

I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled

#### ELECTROLUMINESCENT DEVICES WITH ELECTRODE PROTECTION

the specification of which was filed January 25, 1999 and granted U.S. Serial No. 09/230,401.

I hereby state that I have reviewed and understand the contents of the above identified specification, including the claims, as amended by any amendment referred to above.

I acknowledge the duty to disclose information which is material to the examination of this application in accordance with Title 37, Code of Federal Regulations, §1.56.

I hereby claim foreign priority benefits under Title 35, United States Code, §119(a)-(d) or §365(b) of any foreign application(s) for patent or inventor's certificate, or section 365(a) of any PCT International application designating at least one country other than the United States listed below and have also identified below any foreign application for patent or inventor's certificate or PCT International application having a filing date before that of the application on which priority is claimed:

Prior Foreign PCT International Application(s) and any priority claims under 35 U.S.C. §§119 and 365(a),(b):

|                | •                             |                   | Priority<br>Claimed |     |
|----------------|-------------------------------|-------------------|---------------------|-----|
| PCT/GB97/02039 | PCT                           | 29 July 1997      | (x)                 | [ ] |
| (Number)       | (Country-if PCT, so indicate) | (DD/MM/YY Filed)  | YES                 | ИО  |
| GB_9615883.7   | Great Britain                 | 29 July 1996      | [x]                 | []  |
| (Number)       | (Country)                     | (DD/MM/YY Filed)  | YES                 | МО  |
| GB 9624707.7   | Great Britain                 | 28 November 1996  | [x]                 | ( ) |
| (Number)       | (Country)                     | (DD/MM/YY Filed)  | YES                 | ИО  |
| GB 9703172.8   | Great Britain                 | 15 February 1997  | (x)                 | []  |
| (Number)       | (Country)                     | (DD/MM/YY Filed)  | YES                 | МО  |
| GB 9619382.6   | Great Britain                 | 17 September 1996 | (x)                 | []  |
| (Number)       | (Country)                     | (DD/MM/YY Filed)  | YES                 | МО  |

I hereby claim the benefit under Title 35, United States Code, \$119(e) of any United States provisional application(s) listed below:

| (Application Number) | (filing date) |
|----------------------|---------------|
| (Application Number) | (filing date) |

I hereby claim the benefit under Title 35, United States Code, \$120 of any United States application(s), or \$365(c) of any PCT International application(s) designating the United States of America listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States or PCT International application in the manner provided by the first paragraph of Title 35, United States Code, \$112, I acknowledge the duty to disclose material information as defined in Title 37, Code of Federal Regulations, \$1.56 which became available between the filing date of the prior application and the national or PCT International filing date of this application:

| (Application No.) | (filing date) | (status-patented, pending, abandoned) |  |
|-------------------|---------------|---------------------------------------|--|
|                   |               | •                                     |  |

PCT International Applications designating the United States:

(PCT Appl. No.) (U.S. Ser. No.) (PCT filing date) (status-patented, pending, abandoned)

I hereby appoint the following attorney(s) and/or agent(s) to prosecute this application and to transact all business in the Patent and Trademark Office connected therewith:

Therese A. Hendricks, Reg. No. 30,389 M. Lawrence Oliverio, Reg. No. 30,915

Address all telephone calls to Therese A. Hendricks at telephone no. (617) 720-3500. Address all correspondence to

Therese A. Hendricks
c/o Wolf, Greenfield & Sacks, P.C.,
Federal Reserve Plaza
600 Atlantic Avenue
Boston, MA 02210-2211

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

Inventor's signature

Full name of sole or first inventor Karl Pichler

Citizenship Austria

Residence c/o Fed Corporation, Hudson Valley Research Plc,

1580 Route 52

Hopewell Junction, New York 12533

United States of America

Post Office Address Same

Inventor's signature

Full name of second or joint inventor Carl Towns

Citizenship United Kingdom

Residence 193 Silver Street, Crown Cottage

Stanstead Essex CM24 8HB

Great Britain

Post Office Address Same ?

~ ? SS

Date

12 Now 89

# COPY

#### **ASSIGNMENT**

In consideration of One Dollar (\$1.00) and other good and valuable consideration, the receipt of which is hereby acknowledged, we, the undersigned Karl Pichler and Carl Towns, hereby

Sell, assign and transfer to Cambridge Display Technology Limited, a corporation having a place of business at 181A Huntingdon Road, Cambridge, CB3 ODJ, United Kingdom, its successors, assigns and legal representatives, all hereinafter referred to as the Assignee, the entire right, title and interest for the United States and all foreign countries, in and to any and all inventions which are disclosed in the application for United States Letters Patent Application No. 09/230,401 filed January 25, 1999 and entitled ELECTROLUMINESCENT DEVICES WITH ELECTRODE PROTECTION, and in and to said application and all divisional, continuing, substitute, renewal, reissue and all other applications for Letters Patent which have been or shall be filed in the United States and all foreign countries on any of said inventions; and in and to all original and reissued patents which have been or shall be issued in the United States and all foreign countries on said inventions including the right to apply for patent rights in each foreign country and all rights to priority.

Agree that said Assignee may apply for and receive Letters Patent for said inventions in its own name; and when requested, without charge to but at the expense of said Assignee, agree to carry out in good faith the intent and purpose of this assignment by executing all divisional, continuing, substitute, renewal, reissue, and all other patent applications on any and all said inventions, by executing all rightful oaths, assignments, powers of attorney and other papers, by communicating to said Assignee all facts known to me relating to said inventions and the history thereof, and generally by doing everything possible which said Assignee shall consider desirable for aiding in securing and maintaining proper patent protection for said inventions and for vesting title to said inventions and all applications for patents and all patents on said inventions, in said Assignee.

Hereby request the Honorable Commissioner of Patents and Trademarks to issue said Letters Patent to said Assignee.

Covenant with said Assignee that no assignment, grant, mortgage, license or other agreement affecting the rights and property herein conveyed has been made to others by me, and that full right to convey the same as herein expressed is possessed by me.

Dated: 12 12 8 99

Karl Pichler

Dated: 5.399.

CRUS



PATENT Customer No. 22,852 Attorney Docket No. 8513-7005

#### IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

| n re Application of:                                       |                           |
|--|---------------------------|
| Karl Pichler et al.  | Group Art Unit: 2812      |
| Serial No.: 09/230,401                                     | )<br>Examiner: S. Mulpuri |
| Filed: January 25, 1999                                    |                           |
| For: Electroluminescent Devices With Electrode Protection  | )<br>)                    |
| Assistant Commissioner for Patents<br>Washington, DC 20231 |                           |
| Sir:   |                           |

#### **CHANGE OF CORRESPONDENCE ADDRESS**

Effective immediately, please address all future correspondence with respect to the above-identified patent application to the following address:

FINNEGAN, HENDERSON, FARABOW, GARRETT & DUNNER, L.L.P.
1300 I Street, N.W.
Washington, D.C. 20005-3315
(202) 408-4000 (Telephone)
(202)-408-4400 (Facsimile)

Respectfully submitted,

FINNEGAN, HENDERSON, FARABOW, GARRETT & DUNNER, L.L.P.

Dated: August 7, 2001

M. Lawrence Oliverio

Reg. No. 30,915